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Microwave Assisted Dipole-Dipole Transitions

by

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April 24, 2017

Submitted to the Faculty of Ursinus College in fulfillment of
the requirements for Honors in the Department of Physics.

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Abstract

We explore this two photon assisted transition through computational and numerical analysis of possible energy levels. We calculate the matrix elements of the energy transition in detail discussing constants and the quantum mechanical possibilities of energy exchanges in these systems.

The goal is to better understand the energy exchange, so that moving forward we can control it. This paper covers the theoretical ends to controlling the energy transition by the way of two photon assisted transitions. The energy transitions take place between a dipole-dipole interaction, and a microwave photon.

Acknowledgments

I would like to thank Tom Carroll not only for his guidance in this research, but for his role as a professor, mentor, and friend. He has changed my perspective on science and life, and continues making a difference in students lives everyday.

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Chapter 1

Introduction

Quantum mechanics is the fundamental theory of nature and is particularly necessary at atomic and subatomic scales. Some Quantum mechanical systems for example solid state systems are complex and difficult to study. One method to study such quantum mechanical systems is quantum analog modeling. Quantum analog modeling uses simple quantum systems that can be controlled and easily constructed, and applying the results to more complex quantum systems [2]. The control of quantum mechanical systems extends into quantum computing, systems of atoms can be used to turn long range interactions on and off [3]. By using a system of quai-one-dimensional atoms it is possible to control the statistical coherence through electron wave packets [4, 5]. It is also possible to control highly excited electrons through genetic algorithms [6]. These levels of control are motivation to further study controlling quantum mechanical systems.

Quantum mechanical systems can be studied in numerous branches of physics, but we focus on studying these system on the atomic level. In our experiments we trap the atoms to study them. A trap is used to cool atoms to micro-kelvin, or ultra-cold, temperatures. Since the atoms are ultra-cold they move very slowly or close to not at all on experimental timescales of 10s of μs. Slower moving atoms means there will be fewer collisions allowing the study of long range interactions.

Since they are practically frozen they are easy to probe and control with lasers.

There are various types of atomic traps, however the one used in experiments in this paper used a magneto-optical trap, or MOT. The MOT uses a pair of anti-Helmholtz coils to create a magnetic field around a vacuum which houses the atoms [7]. The atoms are cooled to ultra-cold temperatures using three propagating lasers, these lasers force the atoms to stay in the intersection of the beams.

Controlling quantum interactions has long been a goal in the field of atomic physics. To control the system means there are "knobs", "switches", or "buttons" that can be modified to study a system in numerous ways. One method is to adjust the electric field strength to shift the energy levels. One device can not be controlled with the control of a single parameter. To create simulations that accurately describe the system and aid in the understanding of more complex systems we need multiple knobs, switches, and buttons.

The system in this paper has already achieved one "knob" from previous work [1]. This previous work studied the angular dependence of the system. This angular dependence was controlled using the orientation of the electric field. We were able to conclude that the system did indeed depend on the orientation of the atoms in the system, and now provides the means for controlling the system. Just by literally using a knob, we are now able to use the electric field orientation and strength to control the system. Prior to this the knobs have been an idea of controlling a parameter theoretically. This knob that controls the electric field is actually a knob and we can turn it to increase the electric field and turn it the other way to decrease the electric field. We are also able to use a device to change the direction of the electric field. The process will be discussed more in section (1.5). It is important to note that though these ideas at the moment are focused on studying theoretical

knobs, buttons, and switches, but are able to be constructed to be used in both simulations and experiments.

The goal now is to add another knob, switch, or button to the system to help control the system to a higher degree. Moving forward we are able to look at previous works in configuration interaction models that study coupled states [8, 9]. Currently we are studying the two photon transitions assisted by microwave radiation on the atomic level Using microwave radiation allows us to flip a switch and have the interactions take place, or not take place. This idea is known as a two photon interaction. This interaction does not occur naturally without the microwave radiation. This means we are able to control the interaction using a switch to stop the microwave radiation from entering the system, and at the same time turn it on when needed. This level of control is what is needed to better understand the quantum mechanical system.

1.1 Review of Notation

There are many atomic levels including but not limited to an s-state, p-state, and d-state. These states have specific quantum numbers that describe their properties. The principle quantum number n, refers to the energy level of the state, in Fig. 1.3 the n values are clearly labeled for the given states being studied. There is also the angular quantum number ℓ specifies the shape of the orbital. Lastly the magnetic quantum number m specifies the orientation in space [10]. where j is the total angular quantum number, ℓ corresponds to the rotational angular momentum $s = 1$, $d = 2$, and $f = 3$, and m_j, the magnetic quantum number, is $-j, -j + 1/2, ..., j - 1/2, j$.

1.2 Rydberg Atoms

The atoms we utilize in the system are known as Rydberg atoms. The Rydberg atoms are created using a group I element. Group I elements have a single valence electron. The valence electron is loosely bound because it is in an energy level by itself. Since the electron is loosely bound we are able to more directly add energy to that level and excite the the electron to a Rydberg state. To excite this electron we use a laser to provide it with energy to transition to a significantly higher energy level than it would be at naturally.

After the atoms are trapped in the MOT, a laser is used to excite the valence electron of the atoms to a Rydberg state. This Rydberg state is excited to a much higher energy level than the rest of the electrons in the system. In Fig. 1.2 is a simple model of an atom that has transitioned into a Rydberg atom. In this Bohr model the hydrogen atom has a single proton with charge $+e$, and an electron with charge $-e$. The Bohr model of hydrogen can be compared to a Rydberg atom, with the Rydberg atom there is a positive core with charge $+e$ and a single electron with charge $-e$ that has been separated a large distance from the core.

There are advantages to using a system that involves Rydberg atoms. A Rydberg atom is simple enough that it can be modeled like Bohr's hydrogen model, but complex enough interesting things happen on the atomic scale. The electron, after being excited to a Rydberg state, is easily affected by the electric field and can now be finely perturbed. The atom itself is still neutral and thus interacts very little with the environment. These properties make Rydberg atoms attractive for studying quantum mechanical systems.

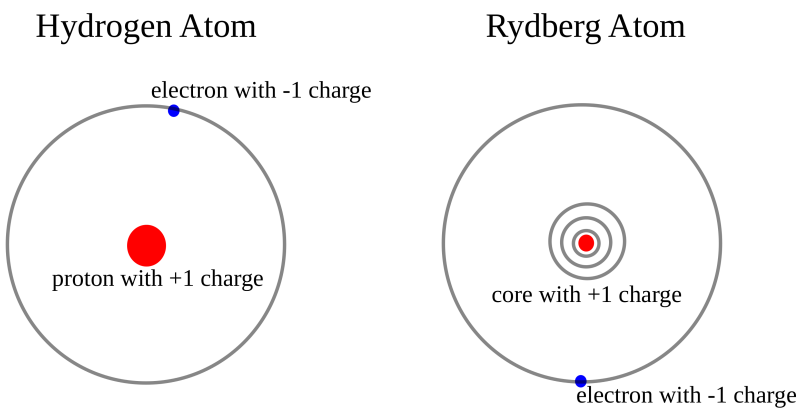

Figure 1.1: A comparison between the Bohr model of a hydrogen atom, and a Rydberg Atom. On the left is the model of the hydrogen atom with a proton being orbited by electron. On the right is a model of an atom that has had its' valence electron excited into a Rydberg state. This Rydberg atom has a positive core with charge $+e$, and an electron.

1.3 Stark Effect

Rydberg atoms are highly sensitive to electric fields and thus thus their energy levels will be strongly perturbed by the Stark effect. The Stark effect causes the energy levels of the Rydberg electron to change as the applied electric field varies. This causes one state to bend take contributions from surrounding states. When the electric field is zero, the states are true to their labels, and take no contributions from neighboring states.

In Fig.1.3 there are two labeled energy states, $39p_{3/2}$ and $39s_{1/2}$. On the vertical axis is the energy of the states, the closer to zero we get the higher the energy level, and the further from zero the more ionized the state is. On the horizontal axis is the strength of the electric field. There is a noticeable bending of the $39p_{3/2}$ energy as the electric field increases.

This bending is the state taking on some contribution from the $n = 36$ state labeled in Fig. 1.3 and becoming a superposition of the two states.The bending that is taking place will factor in to calculating the matrix elements for our system. For example, consider an electron 75% in state A, and 25% in state B, then there are significant contributions from the energy levels of both states. We would have to say this electron is in a superposition of A and B and thus the calculation for the matrix element will have to account for this.

1.4 Dipole-Dipole Interactions

Consider two Rydberg atoms separated by a distance R as shown in Fig. 1.2. Rydberg atoms possess large dipole moments, making interactions between them dipole-dipole interactions. Call the first atom A, and the second atom B. Fig. 1.2 the Ryd-

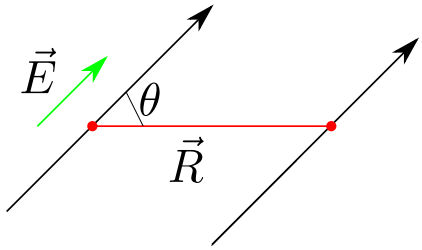

Figure 1.2: Model of dipoles with each black line being a dipole with an orientation at angle θ, separated by a distance R. The electric field, $\vec{E}$ in this case is aligned with the same orientation as the dipoles and is marked in green.

berg electron of atom A is dropping down from the initial state and thus emitting a photon with a specific energy equal to the difference of the initial and lower energy levels. The Rydberg electron of atom B absorbs the Rydberg electron of atom A's photon and thus gains enough energy to transition to a new higher energy state.

When this dipole-dipole interaction takes place it is between two separate Rydberg atoms. Each of the Rydberg atoms is separated by a distance R as seen in Fig. 1.2. The electric field is present in the interaction and in Fig. 1.2 is aligned with the orientation of the dipoles which are at an angle θ. This dipole interaction is

$$V(r) = \frac{\mu_1 \cdot \mu_2 - 3(\mu_1 \cdot \hat{R})(\mu_2 \cdot \hat{R})}{R^3}, \tag{1.1}$$

where R is the separation of the atoms, and μ_1, μ_2 are the matrix elements connecting the initial and final states of each atom [1].

With the dipole-dipole interaction it is possible for two different electrons of different energy levels to switch energy levels. It is also possible for two Rydberg electrons to start in the same energy level and for one to emit a photon transitioning to a lower energy level, and for the other to absorb the photon transitioning to a higher energy level. This can happen in two different ways. The first being that the energy the first electron emits, is the right amount for the second electron to

absorb and transition to a higher level. This is a resonant dipole-dipole interaction. The other possibility is that the energy the first Rydberg electron emits is either too little or too much energy for the second Rydberg electron to land in an energy level, and thus requires extra energy. This is known as a non-resonant dipole-dipole interaction.

1.5 Anisotropy of the Dipole-Dipole Interaction

Our previous worked studied on the angular dependence of the energy exchange among Rydberg atoms [1]. In Fig. 1.2 there is an angle θ that shows the orientation of the dipole. Using a resonant interaction as seen in Fig. 1.3, the angular dependence of the system can be studied more closely. The system can be aligned at different values of θ by changing the direction of the electric field. We calculated the dipole dipole coupling including its dependence on the angle θ. We can use the matrix elements we calculated to create simulations of the exchange at different directions of the electric field.

In the simulations to study the angular dependence a disc of atoms was configured with a small group of p atoms surrounded by s atoms as seen in Fig. 1.4. The more red indicates more p atoms, and the blue indicates s atoms. In the simulation it is possible to change the number of both p and s atoms, as well as change the geometry of the atoms. By changing the ratio of atoms the system can behave differently. The goal of the simulation was to determine if the spread of the p atoms over time depended on the orientation of their dipoles.

From Fig. 1.4 we can see that the work resulted in a visible angular dependence. That is the p atoms spread differently depending on the orientation of the electric field. In Fig. 1.4 (a) the orientation of the electric field is in the x direction, relative

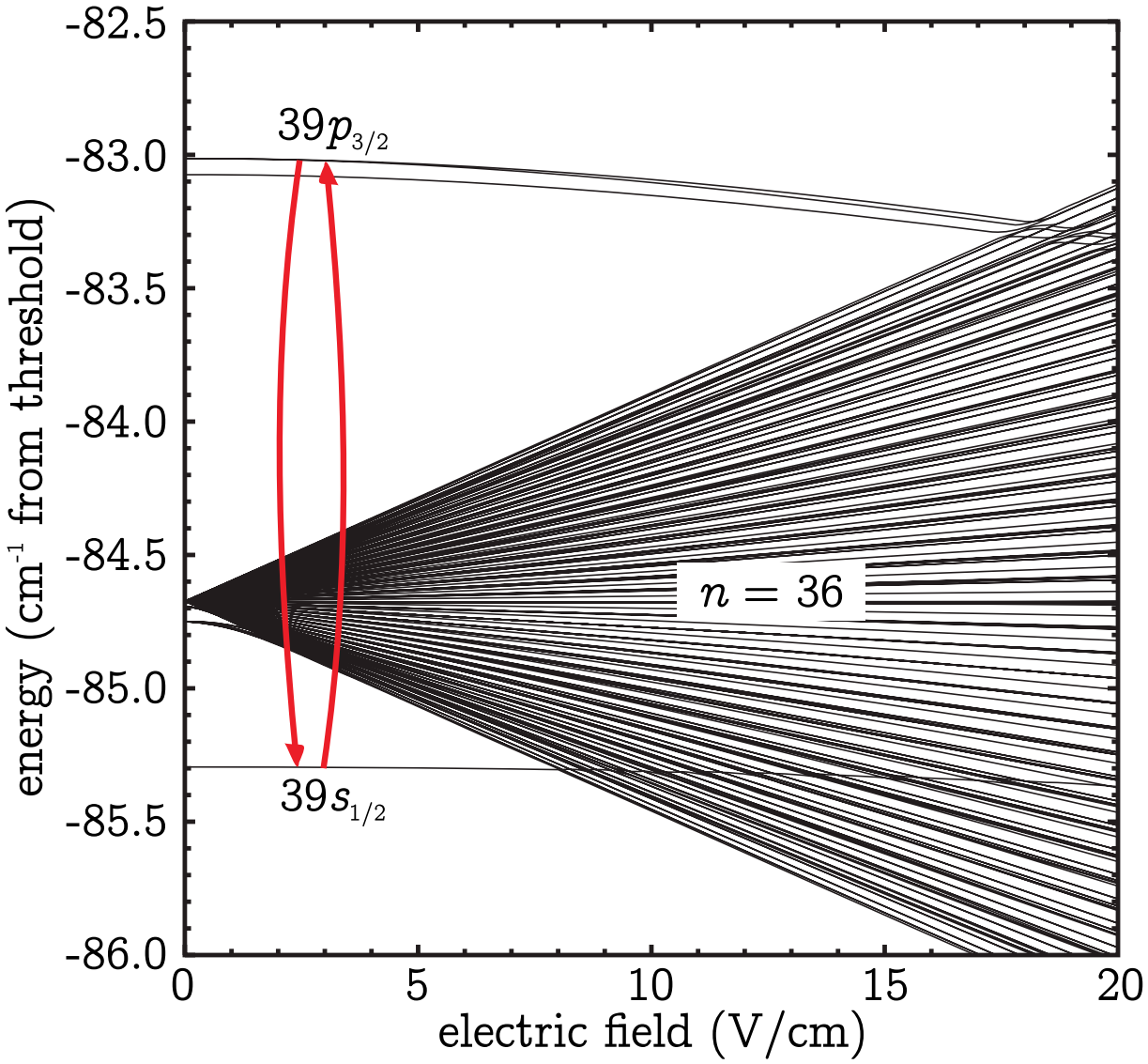

Figure 1.3: A Stark map showing a dipole-dipole interaction $s+p \rightarrow p+s$. This energy exchange is always resonant, meaning that energy that one of the Rydberg electrons emits, is enough for the other to absorb and swap energy levels.

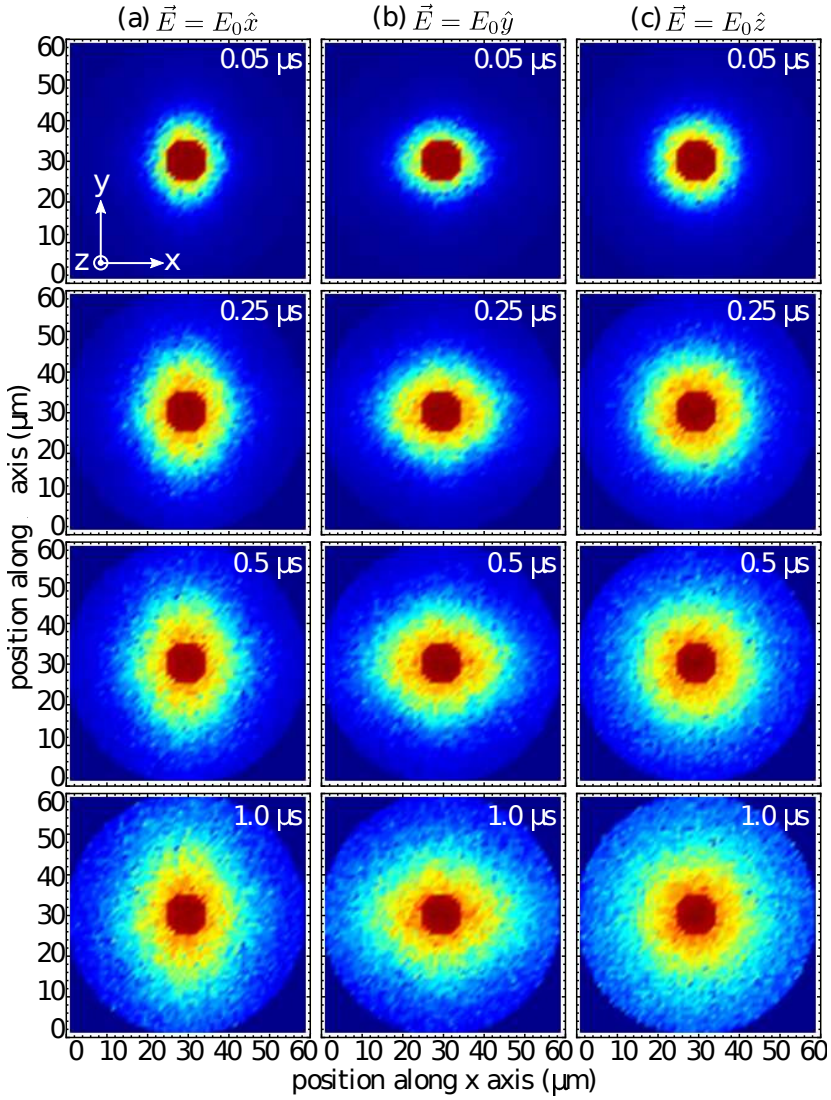

Figure 1.4: The expansion of p character form a centralized disc of atoms. The more red the area the more p probability. The orientation of the geometry it given in the top left corner of the figure. Time progresses downward vertically. (a) The expansion when an electric field $E_0\hat{x}$ is applied. (b) The expansion when an electric field $E_0\hat{y}$ is applied. (c) The expansion when an electric field $E_0\hat{z}$ is applied. In (a) the diffusion of p character is faster in the y direction, while in (b) it is faster in the x direction. Reproduced from [1].

to the axis labeled in the figure. As time increases the p atoms spread is more apparent in the y direction. In Fig. 1.4 (b) the electric field is in the y direction. As time progresses the p atoms are spreading more predominately in the x direction. A reference for an even distribution can be seen in Fig. 1.4 (c), where the electric field is being applied in the z direction. Here the spread is evenly distributed on the disc, because we can imagine the disc is a two-dimensional object and instead of spreading off of the disc it is spreading evenly in both x, and y directions.

The results of these simulations show the system can be altered by altering the angle θ between the dipoles. Before the simulation the only control in the system was the geometry and amount of atoms. This angular dependence now provides a way for the system to be controlled, and gives us a "knob" to turn and adjust the spread of energy in the system.

1.6 Radiatively Assisted Energy Exchange

(we want this interaction, because we can use this fact to solve, an opportunity we are taking advantage of)

The goal of this paper is to engineer an interaction that has insufficient energy from a dipole-dipole interaction and provide it with the energy it needs to happen. To do this we need to focus on interactions that can not take place without added microwave radiation. Consider an interaction like the one shown in Fig. 1.5, recall that a dipole-dipole interaction occurs when one Rydberg electron emits a photon, and another Rydberg electron absorbs that photon, but there is the possibility that this interaction does not happen because the energy the Rydberg electron absorbs is too much, or too little energy to reach an energy level. However we can solve this by adding the microwave radiation, in this case depicted as the blue line in the

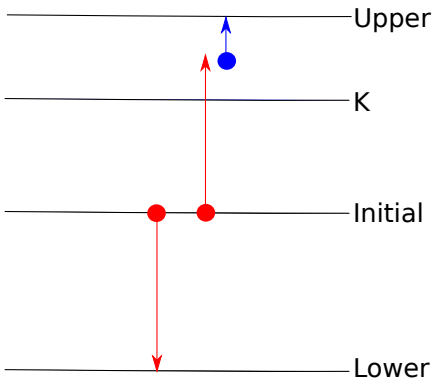

Figure 1.5: A simple model of a possible interaction, the red lines represent atoms undergoing the dipole-dipole interaction while the blue represents a microwave photon. The lines represent the initial, lower, upper, and intermediate energy level K.

figure. We use microwave photons because they are approximately the same scale we need for studying atomic physics.

Now that more than one photon is being absorbed we must calculate new matrix elements to study the system. Quantum mechanics says that every transition that can happen will have a probability amplitude, so to solve for the matrix elements we must sum over all possible cases. One of these sums has to account for the fact that the dipole-dipole interaction can happen first followed by the microwave photon, or that the microwave photon affects the system first followed by the dipole-dipole interaction.

The goal of studying radiatively assisted energy transitions is to add another knob, switch, or button to our system. The system will be able to have different interactions with larger gaps in the energy levels with the flip of a switch. Once the matrix elements are calculated, the simulations can be created. Using simulations allows for guidance in experimental work. If we can properly calculate the matrix elements and create significant simulations than it gives the experimental work a starting point. Once the simulations work preliminary data can be taken. This data

can be used to guide our experiments, for example we can use the simulations to find which states more probable transitions. We will also be able to learn how much microwave radiation is needed at specific strengths of the electric field. Overall the calculations of these two photon transitions make it possible to create simulations with parameters we can control to collect preliminary data for specific experiments.

Chapter 2

Theory

To better understand the process of calculating matrix elements of possible energy exchanges we will walk through multiple examples including finding Clebsch-Gordan Coefficients, and calculating matrix elements for a two photon transition. The system is a quantum mechanical system, and thus needs to account for all the possibilities. For a two photon transition it is possible for the Rydberg electron to pass through a virtual intermediate state that has many substates. If this is the case it is required to sum over all of these states and account for them in the integral.

To show the calculation for one of the possible states, we will look at the case of an atom transitioning from the $42d_{5/2}$ state to the $44p_{5/2}$ state. This example will give us an opportunity to look at both d and p states with different j values. We will be using bra-ket notation with the notation, $\langle j_{final}, \ell_{final}, m_{j-final} | j_{initial}, \ell_{initial}, m_{j-initial} \rangle$. For this problem we will be using $m_j = 1/2$. For the angular interaction we use the operator er. However to get a numerical value for examination we must change j and m_j to ℓ and m_ℓ respectively, along with introducing an m_s term. To do this we need to change basis, and find the Clebsch-Gordan coefficients for the givens states. To change a bases we need to understand the relationship between the two different states and how they are different [10].

The states we start with can be written as a superposition or combination of

the state we wish to change to. This means one of our starting states will become a superposition state in our new basis. We start with the uncoupled basis of $|j_1 m_1; j_2, m_2; M\rangle$ and move to a coupled basis where the eigenfunctions are $|j_1, j_2, J, M\rangle$ [11].

2.1 Matrix Elements for a Photon Transition

To create simulations it is required to solve for the matrix elements of a photon transition. There are two cases to consider, the first being a single photon transition, and the second being a two photon transition. The single photon transition is a resonant interaction where one photon is able to allow the transition of two Rydberg electrons. A two photon transition is the non-resonant interaction, where a second photon is needed for one of the transitions.

2.1.1 Matrix Elements for a Single Photon Transition

A single photon transition is a resonant interaction. This means that one Rydberg electron is able to transition to a lower energy level emitting a photon of a certain energy, which we can call ω_d. Another Rydberg electron is able to absorb the emitted photon transition to a higher energy level, ω_d energy.

Part of the matrix element can be described as,

$$M_{i\ell} = \langle lower|r|initial\rangle \,. \tag{2.1}$$

This matrix element describes the Rydberg electron that is emitting a photon, starting in the initial state i, and transitioning to the lower state ℓ. The other half

of the full matrix element for the dipole-diple interaction is

$$M_{if} = \langle final|r|initial\rangle \,. \tag{2.2}$$

Where i is the initial state and f is the final state. This is the case where a photon is absorbed by the Rydberg electron and transitions to a higher energy state.

It is important to note that the energy from the transition from initial to lower is the same as the energy of the transition from initial to final. In our calculations we control the initial and lower states, calculate the energy, ω_d, for the interaction, and apply that number to the initial state. This will tell us what the final state of the transition is. In other words it is ω_d higher in energy than the initial state.

2.2 Clebsch-Gordan Coefficients

Clebsch-Gordan coefficients arise from the coupling of angular momentum states. Classically when we combine vectors the output is another vector, but when we combine angular momentum vectors we create a new net angular momentum. With our example for this problem we have an atom in a d-state transitioning to a p-state. In Fig. 2.1 we see a small section of the Clebsch-Gordan table focusing on coupling $\ell = 2$ to sping $\frac{1}{2}$ section. In our example , our initial state has $l = 2$ and spin-1/2 electron.

The energy level for this example is $42d_{5/2}$ and $m_j = +\frac{1}{2}$ so we look at the red column,

$$\begin{bmatrix} j \\ m_j \end{bmatrix} \Rightarrow \begin{bmatrix} 5/2 \\ 1/2 \end{bmatrix} . \tag{2.3}$$

Using this method will factor our state into $C_1 \langle \ell, m_{\ell 1}, m_{s1}| + C_2 \langle \ell, m_{\ell 2}, m_{s2}|$. These

coefficients are found in the table. The variables $m_{\ell 1}$ and m_{s1} represent the the two top numbers in the green box in Fig. 2.1, $+1$, and $-1/2$ respectively. To the right of these numbers is a blue box the top blue box is the Clebsch-Gordan coefficient that arrives from this basis change, giving us $C_1 = \sqrt{2/5}$. All Clebsch-Gordan coefficients are the square root of the value given on the table. Looking at the lower green and blue boxes we get $m_{\ell 2}$ and m_{s2} are 0 and $+1/2$ respectively. To the right of this set is another Clebsch-Gordan coefficient, $C_2 = \sqrt{3/5}$. We started with $\langle j = 5/2, \ell = 2, m_j = 1/2|$ and changed our basis to

$$\sqrt{\frac{2}{5}}\,|l = 2, m_{\ell 1} = 1, m_{s1} = -1/2\rangle + \sqrt{\frac{3}{5}}\,|\ell = 2, m_{l2} = 0, m_{s2} = +1/2\rangle\,. \tag{2.4}$$

For simplicity we can drop the quantum number labels ℓ m_ℓ, and m_j and write Eq. 2.4 as,

$$\sqrt{\frac{2}{5}}\,|2, 1, -1/2\rangle + \sqrt{\frac{3}{5}}\,|2, 0, +1/2\rangle \tag{2.5}$$

The initial basis has to be changed as well. We start with our sate as $|j = 3/2, \ell = 1, m_j = +1/2\rangle$. Proceeding as in Eq. (2.4)-Eq. (2.5) we look at Fig. 2.2 which is a section of the table $1 \times 1/2$ which refers to $\ell = 1$ and the spin being 1/2. This time want to look at the column

$$\begin{bmatrix} 3/2 \\ +1/2 \end{bmatrix}, \tag{2.6}$$

outlined in red. From here we look at the two new states in green where $m_{\ell 1} = 1$, $m_{s1} = -1/2$, $m_{\ell 2} = 0$, and $m_{s2} = +1/2$. The blue box now gives us Clebsch-Gordan

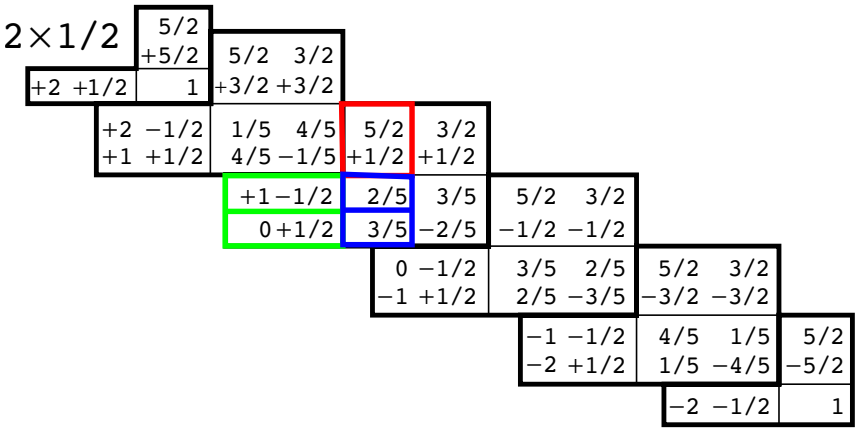

Figure 2.1: A small section of a Clebsch-Gordan table of coefficients focusing on coupling $\ell = 2$ and spin $\frac{1}{2}$. The red box indicates the relevant state, the top green box represents the first part of the basis change where the first two numbers are the new m_l, and m_s respectively. The bottom green box represents the other part of the state from the basis change. The blue boxes represent the Clebsch-Gordan coefficients that correspond to each new state. In this image we are using a $j = \frac{5}{2}$, $m_j = +\frac{1}{2}$.

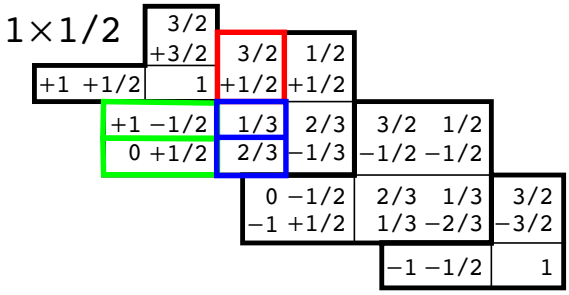

Figure 2.2: A small section of a Clebsch-Gordan table of coeffecients focusing on coupling $\ell = 1$ and spin $\frac{1}{2}$. The red box indicates the relevant state, the top green box represents the first part of the basis change where the first two numbers are the new m_ℓ, and m_s respectively. The bottom green box represents the other part of the state from the basis change. The blue boxes represent the Clebsch-Gordan coefficients that correspond to each new state.In this image we are using a $j = 3/2$, $m_j = +1/2$.

coefficients $C_1 = \sqrt{1/3}$, and $C_2 = \sqrt{2/3}$. Now our final state becomes

$$\sqrt{1/2}\,\langle \ell = 1, m_{\ell 1} = 1, m_{s2} = -1/2| + \sqrt{2/3}\,\langle \ell = 1, m_{l2} = 0, m_{s2} = +1/2| \quad (2.7)$$

for simplicity we drop the variables, and can write Eq. 2.7 as,

$$\sqrt{1/3}\,\langle 1, 1, -1/2| + \sqrt{2/3}\,\langle 1, 0, +1/2| \quad (2.8)$$

By combining both our new states we get,

$$\left(\sqrt{\frac{1}{3}}\,\langle 1, 1, -1/2| + \sqrt{\frac{2}{3}}\,\langle 1, 0, 1/2|\right)(er)\left(\sqrt{\frac{2}{5}}\,|2, 1, -1/2\rangle + \sqrt{\frac{3}{5}}\,|2, 0, 1/2\rangle\right) \quad (2.9)$$

This product now has to be multipled out, to identify each term better we will break it up into parts.

$$\begin{aligned}\sqrt{\frac{1}{3}}\,\langle 1, 1, -1/2|\,(er)\sqrt{\frac{2}{5}}\,|2, 1, -1/2\rangle + \sqrt{\frac{1}{3}}\,\langle 1, 1, -1/2|\,(er)\sqrt{\frac{3}{5}}\,|2, 0, 1/2\rangle + \\ \sqrt{\frac{2}{3}}\,\langle 1, 0, 1/2|\,(er)\sqrt{\frac{2}{5}}\,|2, 1, -1/2\rangle + \sqrt{\frac{2}{3}}\,\langle 1, 0, 1/2|\,(er)\sqrt{\frac{3}{5}}\,|2, 0, 1/2\rangle \quad (2.10)\end{aligned}$$

Two of the terms, the second and third, have differing m_ℓ values, this is not possible by selection rules and thus are equal to zero [12]. Moving forward we need to integrate the terms. The terms can be broken into a radial and angular integral,

$$e\int_0^{2\pi}\int_0^{\pi} Y_{\ell_f, m_f} Y_{\ell_i, m_i} d\Omega \int_0^{\infty} R_{42d_{5/2}} R_{44p_{3/2}}(r) r^2 dr. \quad (2.11)$$

The first set of integrals are spherical harmonics that cover the angular part of the integration, the second integral is the radial part of the equation. The integration

of the angular part yields,

$$\sqrt{\frac{\ell^2 - m_\ell^2}{(2\ell+1)(2\ell-1)}} \tag{2.12}$$

[13]. This equation uses the initial ℓ value for the interaction, in our case that would give us $\ell = 2$, and $m_\ell = 1$. We can evaluate both angular terms. For the first surviving case we get,

$$\sqrt{\frac{\ell^2 - m_\ell^2}{(2\ell+1)(2\ell-1)}} = \sqrt{\frac{3}{15}} \tag{2.13}$$

and for the later,

$$\sqrt{\frac{\ell^2 - m_\ell^2}{(2\ell+1)(2\ell-1)}} = \sqrt{\frac{4}{15}}. \tag{2.14}$$

This leaves us with the radial part, this calculation is done using computational software. To calculate this numerical term we use Numerov's method [14]. To calculate Numerov's method for these terms precisely we use the super computer to account for the contribution from other states. As discussed in section 1.3, the states in an electric field are the superposition of the state at zero energy, and the state that it is bending towards on the Stark map as pictured in Fig. 1.3.

Using ξ as the numerical part and pulling out the Clebsch-Gordan coefficients we arrive at,

$$\sqrt{\frac{1}{3}}\sqrt{\frac{2}{5}}\sqrt{\frac{3}{15}}\xi + \sqrt{\frac{2}{3}}\sqrt{\frac{3}{5}}\sqrt{\frac{4}{15}}\xi \tag{2.15}$$

To calculate ξ we look at using computational methods on the super computer. This is because the radial integration in Eq. (2.11) accounts for the states that undergo the bending from the detuning and Stark effect. This means the superposition isn't only a number but rather a sum of integrals.

2.3 Matrix Elements for a Two Photon transition

Two photon transitions are non-resonant interactions as seen in Fig.1.5. This reaction can happen two different ways. The first being that the first Rydberg electron emits a photon transitioning to a lower energy level ℓ with energy ω_d, and the second Rydberg electron absorbs that photon, then the microwave photon provides energy to the system. Here the electron absorbs the same energy as emitted from the first Rydberg electron however due to the non-resonant nature of the interaction, the state passes through a virtual intermediate level k. The virtual level could have multiple substates, and thus has to be summed over. We can call these energies ω_{ki}, the energy from the initial state i to one of the intermediate states k. Another possibility is when the microwave provides energy to the system first, allowing one of the Rydberg electrons to absorb the photon. That interaction is then followed by the dipole dipole interaction where the Rydberg electron that reieved the photon from the radiation recieves another photon from the dipole-dipole interaction. Both of these processes are possible in quantum mechanics and thus both have to be accounted for.

To calculate the matrix elements we write each transition as a sum over all possible interactions. The first Rydberg electron starts in the initial state and transitions to a lower state ℓ, which can be written as $M_{i\ell}$. The other possible transition is between the initial i, and intermediate k levels, and can happen either by dipole-dipole or microwave radiation and is written as M_{ik}. The last transition to take place again happens either between the dipole-dipole interaction or microwave radiation and can be written as M_{kf}.

To create an equation to calculate the two photon transition, we use the terms

for each transition. The summation of these elements yields.

$$\frac{1}{R^3}\left[M_{i\ell}\Sigma\frac{M_{ik}\vec{e_1}M_{kf}\vec{e_2}}{\omega_{ki}-\omega_d}+M_{i\ell}\Sigma\frac{M_{ik}\vec{e_1}M_{kf}\vec{e_2}}{\omega_{ki}-\omega_\mu}\right]. \tag{2.16}$$

Regardless of the interaction we choose $M_{i\ell}$. The electron that emits a photon will always be consistent because when doing the experiments and simulations we will choose which energy level the electron is transitioning to. Since we choose the lower energy level the photon the electron emits, transitioning to the lower energy level, will be known and constant thus we do not have to sum over it allowing us to factor it out of the equation,

$$\frac{1}{R^3}M_{i\ell}\left[\Sigma\frac{M_{ik}\vec{e_1}M_{kf}\vec{e_2}}{\omega_{ki}-\omega_d}+\Sigma\frac{M_{ik}\vec{e_1}M_{kf}\vec{e_2}}{\omega_{ki}-\omega_\mu}\right]. \tag{2.17}$$

In this equation, the M factors represent the matrix elements and their respective transitions, while ω_d is the energy gap between the initial and lower levels, ω_{ki} is the series of gaps between the initial level and sum of virtual intermediate levels, and ω_{kf} is the sum of possible energy levels from the virtual intermediate state k and the final state.

2.3.1 Calculations of ω

The ω values that arise through these transitions are differences between energy levels. Each ω value represents a different energy gap between the states. In Fig. 3.2, ω_d is the energy give by

$$\omega_d = E_i - E_\ell. \tag{2.18}$$

Where E_i is the energy of the initial state and E_ℓ is the energy of the lower state. Another ω term we can calculate is ω_{ki}, the energy between the initial state i, and the virtual intermediate state k. This calculation will be for a chosen value depending on ω_d, but can be described as

$$\omega_{ki} = E_k - E_i. \tag{2.19}$$

It is important to note that the k state can have multiple substates, and thus ω_{ki} can have multiple values. The last ω term in Eq. (2.17) is ω_μ, the last remaining energy needed to reach the final state f. Assuming the Dipole-dipole itneraction happens first we know the energy from the initial level to the lower level is ω_d we also know the second electron in the initial states rises $omega_d$ in energy so we can write our value as

$$\omega_\mu = E_f - 2\omega_d. \tag{2.20}$$

Chapter 3

Results

Calculating examples to test theoretical work is always necessary. In the following chapter we test our method against previous work [8, 9], and complete an example of a system where $\omega_d \approx \omega_\mu$. This means that the photon absorbed from the dipole-dipole interaction is almost the same energy as the microwave photon being introduced to the system.

In the work previously completed by the Gallagher group at the University of Virginia who experimentally and theoretically calculated a coupling using a configuration-interaction model. This model is a different way to study the quantum mechanical system [9]. There studies focus on a series of coupled states that are always separated by certain energy gaps. These gaps are chosen by first selecting the initial state to be at an energy level $nd_5/2$, making their coupled initial state $nd_{5/2}nd_{5/2}$. Their lower level is then chosen to be $(n+2)p_{3/2}(n-2)f$. The final energy level of for this model is $(n+1)d_j(n-2)f$ [8]. With this method their goal was to study the interaction and see if their method of configuration-interaction held true for multiple values of n. Our work differs as in we study two photon transitions, and wish to understand the energy exchange so that we can control it. By only choosing the initial and lower states we are able to further study the interaction that is taking place between the virtual intermediate levels and the final

state. We are looking for these matrix elements to create simulations.

3.1 Comparison to Previous Work

In the previous work Yu et-al. [8], and Lee and Gallagher [9] worked on configuration interaction models and derived [8],

$$V = \langle nd_{5/2} nd_{5/2} | \frac{r_1 r_2}{R^3} | (n+2)p_{3/2}(n-2)f \rangle \times \frac{1}{\Delta} \langle (n+2)p_{3/2}(n-2)f | \vec{r_1} \cdot \vec{E} | (n+1)d_j(n-2)f \rangle \,. \quad (3.1)$$

This equation calculates the coupling of states that are energetically nearby. In Eq. (3.1) $\vec{r}_1$, and $\vec{r}_2$ are the Rydberg electrons positions and $\vec{E}$ is the microwave electric field. For their example they let $n = 42$, which can be used to find their initial, intermediate, lower, and final states. Our goal is to use our method to reach a similar matrix element or coupling.

We would like to use Eq. (2.17) to compute the matrix elements using a two-photon transitions. We start by collecting our states. We choose to also let $n = 42$. This means that $\ell = 40f$, $M_i = 42d_{5/2}$, $M_k = 44p_{3/2}$, and $M_f = 44d_{5/2}$. From the use of closely related energy states the virtual intermediate state has been chosen for us by selection rules [8]. For visual representation of this interaction we can look at Fig. 3.1, this shows one of the quantum mechanical possibilities that can take place during this two photon transition. We can rewrite Eq. (2.17) in braket notation giving us,

$$\frac{{a_0}^3 e^3}{4\pi\epsilon_0 R^3} \langle M_\ell | r | M_i \rangle \left[\sum \frac{\langle M_k | r | M_i \rangle \langle M_f | \vec{E} r | M_k \rangle}{\omega_{ki} - \omega_d} + \sum \frac{\langle M_k | r | M_i \rangle \langle M_f | \vec{E} r | M_k \rangle}{\omega_{ki} - \omega_\mu} \right] \quad (3.2)$$

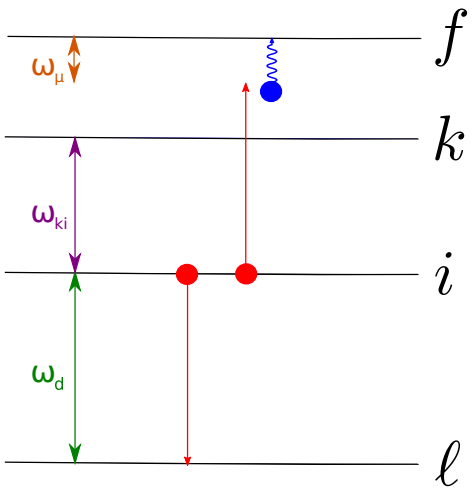

Figure 3.1: This is an updated model of Fig. 1.5. This updated figure revisits the microwave assisted transition. In red is the dipole dipole interaction, and labeled in blue is the microwave photon. ℓ is our lower level, i is the initial level, k is the intermediate level, and f is the final level. ω_{ki} is the energy gap between the initial and intermediate states. ω_d is the energy given up during the dipole dipole interaction and is the gap between the lower energy level and the initial. ω_μ is the energy of the photon being added to the system.

The factor outside of the integration is a constant to keep our answer in SI units. A couple terms arise in this calculation a_0 is the Bohr radius, $\frac{1}{4\pi\epsilon_0}$ is Coulomb's constant, e is the electron mass, and R is the atomic separation of the atoms. Doing this provides us with $\langle 40f|r|42d_{5/2}\rangle = 646ea_0$, $\langle 44p_{3/2}|r|42d_{5/2}\rangle = 628ea_0$, and $\langle 44d_{5/2}|r|44p_{3/2}\rangle = 1154ea_0$. With these numbers we can calculate the ω values in the equation. Using the numbers for the radial integration we get $\omega_{ki} - \omega_d =$ 0.093 GHz, and $\omega_{ki} - \omega_\mu = 40.176$ GHz. Since $\omega_{ki} - \omega_\mu >> \omega_{ki} - \omega_d$ We can say the second term in Eq. (3.2) is approximately zero, yeilding

$$\frac{{a_0}^3 e^3}{4\pi\epsilon_0 R^3} \langle M_\ell|r|M_i\rangle \left[\frac{\langle M_k|r|M_i\rangle \langle M_f||\vec{E}|r|M_k\rangle}{\omega_{ki} - \omega_d}\right]. \tag{3.3}$$

The above equation can be compared to Eq. (3.1). In Eq. (3.1) we can compare our ω values to their Δ value. The product of our sums correlates to their second state,

while our outer state compares to their state unaffected by Δ. The constants we introduced to the term are a correction to SI units, the work in Eq. (3.1) is fo atomic units. Using the values we obtained for the energy levels, and Eq. (3.3), we achieve similar values for the matrix element. This step confirms that our equation works and that we are one step closer to better understanding the two photon transition.

3.2 Calculating Matrix Elements

To construct an example that studies the two photon transition more in depth we choose an example such that $\omega_d \approx \omega_\mu$. This causes the denominator of both terms will be similar and thus not be able to be ignored for our calculations. If we look at Fig. 3.2 it is visible that the microwave photon is almost the same energy as the emitted photon from the dipole-dipole interaction. To study this more we choose a state such that $\ell = 43p$, $i = 44s_{1/2}$, $k = 44p_k$, and $f = 45s_{1/2}$. This gives us,

$$\frac{a_0^3 e^3 |\vec{E}|}{4\pi\epsilon_0 R^3} \langle 43p_k|r|44s_{1/2}\rangle \left[\Sigma \frac{\langle 44p_k|r|45s_{1/2}\rangle \langle 45s_{1/2}|r|44p_k\rangle}{\omega_{ki} - \omega_d} + \Sigma \frac{\langle 44p_k|r|45s_{1/2}\rangle \langle 45s_{1/2}|r|44p_k\rangle}{\omega_{ki} - \omega_\mu}\right]. \quad (3.4)$$

We call the intermediate states a p_k state because there are two possibilities which need to be summed over. This means $k = 1/2$ or $k = 3/2$, both of which are equally possible. This brings up the fact that our ω values differ depending on which state is used. The state does not differ tremendously however. To create a generic formula for solving the above equation we created a piece of code in Mathematica (appendix 1). In this code we used our previous methods of finding the value of the states and created a list to create multiple sums.

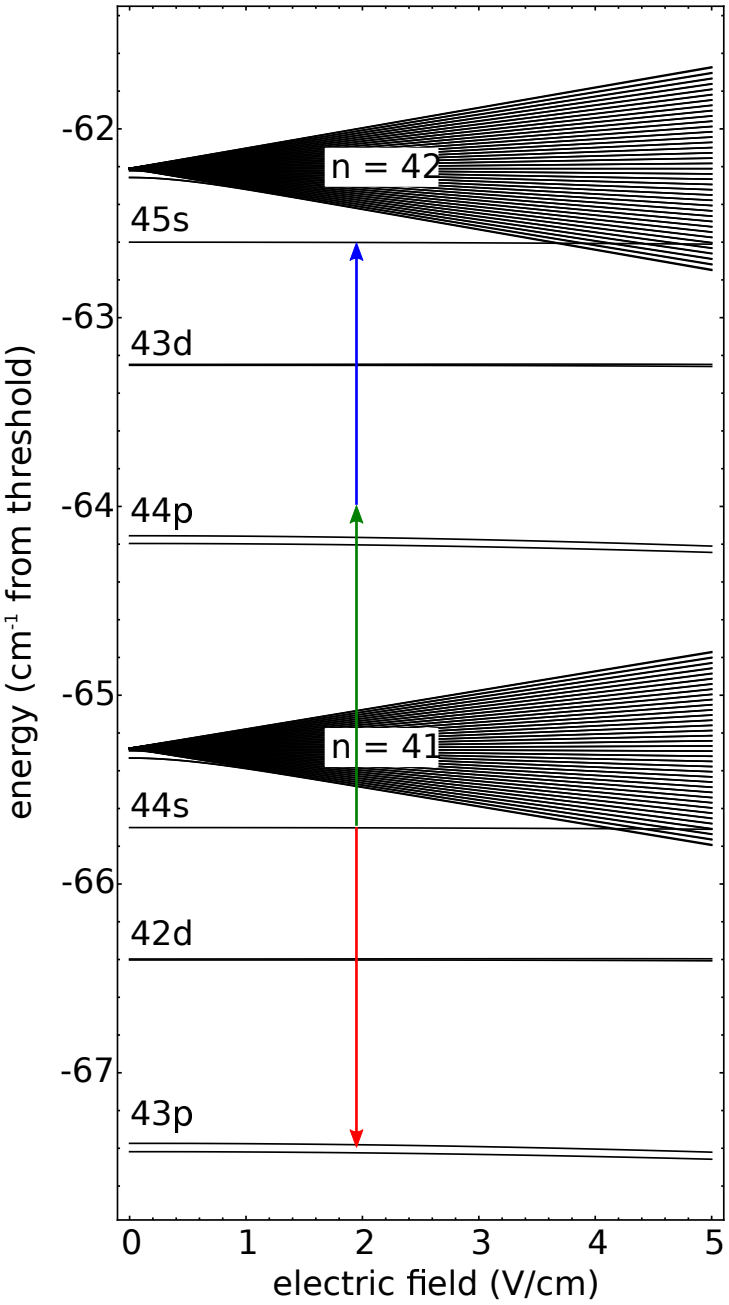

Figure 3.2: A stark map indicating the dipole-dipole interaction of an initial state $44s$, with one Rydberg electron emitting a photon dropping to a $43p$, while the other absorbs the same photon and a microwave photon to reach the $45s$ level. Note that the red and green lines are equal in length and represent the dipole-dipole interaction. The blue line, or microwave photon, is almost as long as the dipole interaction.

Table 3.1: Table consisting of values for energy transitions of particular states. The first column depicts which matrix element is being calculated. The second column provides which state is being tested for different p-states. The last column gives the numerical value calculated using the super computer and Numerov's method.

Matrix Element	States	Values
$M_{i\ell}$	$\langle 43p_{1/2}\|r\|44s_{1/2}\rangle$	$582ea_0$
	$\langle 43p_{3/2}\|r\|44s_{1/2}\rangle$	$838ea_0$
M_{ik}	$\langle 44p_{1/2}\|r\|45s_{1/2}\rangle$	$648ea_0$
	$\langle 44p_{3/2}\|r\|45s_{1/2}\rangle$	$902ea_0$
M_{kf}	$\langle 45s_{1/2}\|r\|44p_{1/2}\rangle$	$611ea_0$
	$\langle 45s_{1/2}\|r\|44p_{3/2}\rangle$	$880ea_0$

To calculate Eq. (3.4), we used the computer code to solve for the energy of our states. The value of all the matrix elements in this calculation can be found in Table. 3.1. The matrix element calculation using $k = p_{1/2}$ gave us 4.2 kHz, while using $k = p_{3/2}$ we got 7.8 kHz. These results are similar to each other and the work provided by Yu et-al. and Lee and Gallagher.

Chapter 4

Conclusion

We were able to create an equation that is capable of creating matrix elements needed for simulations on two photon energy transitions. This equation was tested against previous work and as of now is believed to be a complete calculation of all required matrix elements.

Students in the future will need to use these calculations to create simulations. The simulations should be able to control the initial values, as well as the final state. With the information about those two states, the simulations should be able to run calculations to find a final state and sum over the virtual intermediate states. Once these future simulations can do such calculations they should lead the way to experimental work, and provide preliminary data.

The experimental work will require the careful construction of a system that places microwave radiation inside of the MOT. The microwave radiation needs to be controlled so that the photons being absorbed by the Rydberg electrons in the system can receive the correct energy. Information on the correct energies will come from the simulations.

Once the simulations can match the experimental work we will be able to conceive further work in understanding full control of a two photon transition. This control of two photon transitions will provide another switch will further help the

understanding of the energy exchange among Rydberg atoms, and bring us one step closer to controlling the energy exchange.

There are some assumptions made in our calculations that can be accounted for in future work. We assume the polarizations of the atoms are identical. Shown in our previous work, the orientation of the radiation compared to the dipoles will have to be accounted for.

Appendix A

Mathematica Code

The following code is designed for the input of energy values for an initial , intermediate, and final state. These energy values are received from code on the super computer. These values are then put into the equation in the code, which outputs a numerical answer in hertz. This value is the matrix element for the provided two photon transition.

```
(*Code written for calculating the
  matrix  elements  of a photon assisted transition*)
```

In[130]:=
```
e = 1.6 * 10^-19;
a0 = .529 * 10^-10;
R = 2.4 * 10^-6;
EE = 35 * 10^-3;
e0 = 8.85 * 10^-12;
h = 6.626 * 10^-34;
k = {64.1962, 64.1552};
(*List of values for the intermediate  state, {1/2, 3/2}*)
i = {65.7011};        (*List of values for the initialstate, {1/2}*)
L = {67.4181, 67.3739};  (*List of values for the lower state,
{1/2, 3/2}, correlates to script l in the text.*)
f = {62.6001};        (*List of values for the final state, {1/2}*)
ωj = (Table[Abs[i[[1]] - L[[n]]], {n, 1, Length[k]}]) * (30 * 10^9);
(*A table calculating all possible omega_d  values,
but we choose omega_d  to be one of these*)
ωd = ωj[[2]];
ωki = (Table[Abs[k[[n]] - i[[1]]], {n, 1, Length[k]}]) * (30 * 10^9);
(*To put the wave number  in gigahertz we multiple  by 30*10^9*)
ωp = Table[Abs[f[[1]] - (ωj[[n]] / (30 * 10^9) + i[[1]])], {n, 1, Length[k]}] * (30 * 10^9);
(*A table calculating all possible omega_mu   values,
but we choose omega_mu   to be one of these*)
ωμ = ωp[[1]];
Mik = {648, 902};  (*List of states calculated using
  super computer  first and second correlate to 1/2, and 3/2*)
MiL = {582, 838};
Mkf = {611, 880};
```

In[145]:=
```
1/h ((a0^3 * e^3 * EE * MiL[[2]]) / (4 * π * e0 * R^3)) (Sum[(Mik[[n]] * Mkf[[n]]) / (ωki[[n]] - ωd) * h, {n, 1, Length[k]}] +
    Sum[(Mik[[n]] * Mkf[[n]]) / (ωki[[n]] - ωμ) * h, {n, 1, Length[k]}])
```

Out[145]=
```
-7882.66
```

```
-4189.229882951762`  (*4.2 kHz*)
```

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